Proceedings of the XVI ECSMGE
Geotechnical Engineering for Infrastructure and Development
ISBN 978-0-7277-6067-8

doi:10.1680/ecsmge.60678

Hydraulic conductivity and swelling pressure of GCLs using polymer treated clays to high concentration $CaCl_2$ solutions

Conductivité hydraulique et pression de gonflement de GCLs utilisant des argiles traitées avec polymères à hautes concentrations de $CaCl_2$

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ABSTRACT A Geosynthetic Clay Liner (GCL) is a frequently used hydraulic barrier system designed to impede the flow of contaminated leachate into the environment. The main objective of this barrier system is to maintain a low hydraulic conductivity that is determined by the bentonite fraction. In this study, calcium bentonite, natural sodium bentonites, and sodium activated bentonite were treated with the HYPER clay technique. This involves the adsorption of an anionic polymer, Sodium CarboxyMethylCellulose (Na-CMC) onto the surface of the clay material. The purpose of this research was to show the beneficial effect of the HYPER clay treatment on the swelling and hydraulic performance, while the bentonite is permeated with high concentration $CaCl_2$ solutions. The test results showed that swelling and hydraulic performance increased with Na-CMC treatment, regardless of the type of bentonite that was used. Additionally, a powdered Na-CMC configuration provided higher swelling and hydraulic performance compared to a granular configuration.

RÉSUMÉ Les géosynthétiques bentonitiques (GCL) sont un système de barrière hydraulique souvent utilisés pour empêcher du lixiviat contaminé d'entrer dans l'environnement. Le but le plus important de ce système de barrière est de maintenir une conductivité hydraulique basse, qui est déterminée par la fraction de bentonite. Dans cette étude des bentonites de calcium, des bentonites naturelles de sodium, et des bentonites activées ont été traités selon la méthode de traitement de la HYPER clay. Ceci implique l'adsorption d'un polymère anionique, la CarboxyMethylCellulose (Na-CMC), sur la surface de l'argile. Le but de cette recherche était de montrer l'effet du traitement avec polymères sur la performance hydraulique et le gonflement de l'argile, après contact avec des solutions de $CaCl_2$ de haute concentration. Les résultats montraient que le gonflement et la performance hydraulique augmentaient avec un traitement de Na-CMC, quel que soit le type de bentonite utilisé. En plus, on a trouvé que la Na-CMC en poudre donnait de meilleures propriétés à l'argile que la Na-CMC granulée.

1 INTRODUCTION

With the current consumer society, wastes that are created require disposal despite the best waste management practice. Such wastes include municipal waste, mine tailings, and industrial by-products. When water infiltrates this waste material, contaminated leachate is formed which poses a threat to the environment and human health due to the mobility and solubility of the contaminants in this leachate (Sharma & Reddy 2004). To prevent the spreading of contaminants into the surrounding soil and groundwater, hydraulic barriers are used. These barriers may contain GCLs.

GCLs are factory manufactured clay liners consisting of a thin layer of bentonite supported by geotextiles and/or glued to a geomembrane, mechanically held together by needling, stitching, or chemical adhesives. In present generation landfills, these barriers are used both as liners (at the bottom of the landfill) and covers (on top of the waste).

In these barrier systems, natural calcium or sodium bentonite is used because of their increased sealing capacity in the presence of water. Preferably, sodium bentonite is used because of its higher swelling performance compared to calcium bentonite (Mitchell 1993; Shackelford et al. 2000; Egloffstein 2001). However, sodium bentonite is available to a lesser extent, more expensive, and more prone to cation exchange. Nevertheless, both calcium and sodium bentonite are prone to cation exchange when they are permeated with high concentration solutions (simulation of an aggressive environment), increasing the hydraulic conductivity. This is caused by the collapse of the Diffuse Double Layer (DDL), decreasing the swelling, which causes less tortuous flow paths in the clay structure (Mitchell 1993; Egloffstein 2001). Consequently, a lot of attention must be given to chemical interactions between the clay and contaminated leachate (Mitchell 1993; Petrov & Rowe 1997; Ruhl & Daniel 1997; Jo et al. 2001, 2005; Lee et al. 2005; Katsumi et al. 2008; Scalia et al. 2013).

In order to increase the chemical resistance of clays against aggressive chemicals, modified bentonites have been introduced (Theng 1982; Kondo 1996; Onikata et al. 1999; Xi 2006, Katsumi et al. 2008; Di Emidio et al. 2012; Azzam 2014). In this study, the HYPER clay treatment (Di Emidio 2010) in aggressive environments will be investigated in further detail.

2 MATERIALS

In this study three materials are compared: a natural calcium bentonite (CaB), the same CaB but sodium activated (SACaB), and a natural sodium bentonite; Natural Gel (NG). All of the previous materials will receive the HYPER clay treatment, which consists of mixing the base clay with a polymeric solution (containing a certain percentage of Sodium CarboxyMethylCellulose (Na-CMC) by dry weight of clay) for 30 minutes. Hereafter, the obtained slurry is oven dried at 105 °C for 24 hours to remove all excess water. After drying, the clay is ground manually using a mortar and pestle. Finally, the clay is mechanically ground to a powdered configuration using the Retsch Mortar Grinder RM 200. Some properties of these materials are listed in Table 1.

The electrolyte solutions used in this investigation are deionized water (DI) and $CaCl_2$ solutions with different concentrations. The deionized water was used as base solution for the HYPER clay and electrolyte preparation. The deionized water was produced using the water purification system PURELAB Option-R 7/15. Some properties of the used electrolyte solutions are listed in Table 2.

Table 1. Characteristics of tested soils.

Characteristics	NG	CaB	SACaB
Swell index [ml/2g]	35	12	19
Specific gravity	2.61	2.5	2.5
Liquid limit [-]	583	309.53	414.16
Plastic limit [-]	53	64.03	64.83
Plasticity index [-]	530	245.50	349.33
Smectites [%]	69	82	82
CEC [meq/100g]	83	52	54

Table 2. Chemical properties of the used electrolyte solutions.

Solution	Concentration	EC	Salinity	pH	Eh
	[M]	[mS/cm]	[-]	[-]	[mV]
Deionized water		0.0039	0.0	7.57	293
$CaCl_2$	0.5	78.8	54.3	6.71	187
	0.2	35.7	22.3	7.38	172
	0.1	18.65	11.1	8.02	224
	0.05	9.93	5.6	8.18	230
	0.01	2.27	1.0	6.85	207
	0.005	1.17	0.4	6.65	197
	0.001	0.262	0	7.65	149
	0.0001	0.0416	0	7.76	184

3 METHODS

The tests performed on the three materials were: (1) specific gravity test using 1 gram of bentonite; (2) swell index test using various $CaCl_2$ concentrations; (3) swell pressure test using a 0.005 M $CaCl_2$ solution; and (4) hydraulic conductivity test using a 0.5 M $CaCl_2$ solution. The swell index tests were performed to have a qualitative evaluation of the impact of various electrolyte solutions with different concentrations (0.0001 M up to 0.5 M). However, the swell index test has limitations due to turbidity that occurred during the experiment. In order to overcome these limitations, swell pressure tests were performed on all samples.

The specific gravity of soil solids (G_s) was determined by the water pycnometer method following ASTM D854. The standard states that about 10 g of dry soil per pycnometer should be used. Taking the high swellability of the clay and the high polymer dosage into account, 1 g of soil was used in this study.

Swell index tests were performed following ASTM D5890. 2 g of dry bentonite is added in ±0.1 g increments to 100 ml of test solution in a graduated cylinder. After a minimum hydration period of 16 hours, the final temperature and the volume of swollen bentonite is measured.

The swell pressure test was performed using an apparatus, which consists of a stainless steel ring (10 cm diameter) accommodated in a one-dimensional cell located in a frame provided with a load cell connected to a computer. One swell pressure samples consists of 35.34 g of material. This is calculated by assuming a dry surface density of 0.45 g/cm^2, which is a standard value for GCLs. The bentonite is evenly distributed in a stainless steel ring in between two filter papers and two porous stones. Next, the specimen is permeated with a 0.005 M $CaCl_2$ solution for a minimum of seven days. The loading cell that keeps the height of the sample constant measures the swell pressure.

The swell pressure test has two aims: (1) turbidity or flocculation may occur causing false measurement of the swell index, a swell pressure test is performed to overcome these limitations and to determine the actual swelling performance (Di Emidio 2010); (2) obtaining saturated samples that have a dry surface density which represents a standard GCL core ready to be placed into a flexible wall permeability cell to test its hydraulic conductivity.

3.1 *Hydraulic conductivity test*

The hydraulic conductivity tests were performed in flexible wall permeameters in accordance with ASTM D5084. The tests were executed using a 0.5 M $CaCl_2$ solution with a hydraulic gradient of about 55 and an average effective stress of 35 kPa in a 10 °C room. This effective stress was chosen in order to simulate an extreme condition, such as an average effective stress expected in a landfill cover. In case the effective stress is increased, as it would be the case to simulate a landfill bottom liner, the hydraulic conductivity will decrease accordingly (Petrov & Rowe 1997).

4 RESULTS

This section shows the results obtained from the experimental tests on the clays treated with the HYPER clay technology. Firstly, the specific gravity test results will be shown to determine whether there is a polymer concentration possible where optimum polymer adsorption occurs. Hereafter, swell index results will be shown to have an indication of the impact of electrolyte concentration on treated and untreated clays. According to Jo et al. (2001); Katsumi et al. (2008); Shackelford et al. (2000), there is an inverse relationship between the swell index and the hydraulic conductivity of bentonite clays. Therefore, it can give an idea of the hydraulic conductivity, which could possibly occur in practice. Hereafter, swell pressure test results will be shown. Additionally, Di Emidio et al. (2008); Lee et al. (2012) have shown that there is an inverse relationship between the swell pressure and the hydraulic conductivity. In this way, the swell pressure could give an accurate representation of the hydraulic conductivity, which could occur in practice. Lastly, the hydraulic conductivity results will be shown.

4.1 *Specific gravity by water pycnometer*

Figure 1 shows the results of NG, tested in this study, and a natural sodium bentonite (NaB), tested by Di Emidio (2010). Both of these bentonites have similar characteristics. Considering a polymer concentration ranging from 0 to 8 % for NaB, the specific gravity decreases with increasing polymer dosage. The same was observed for NG in the range of 8 to 10 %. These results can be attributed to several factors, with one of these being the lower specific gravity of the polymer (1.59). Thus, substituting a certain amount of clay for the same amount of polymer will decrease the specific gravity. Another factor is the hypothesis of the polymer maintaining the interlayer between the clay platelets open, limiting aggregation and consequently introducing a dispersed structure. Due to this, the solid fraction will have a larger swell, causing an increase in the total volume, hence a decrease in density and specific gravity. In the range of 8 to 16 % for NaB and 10 to 16 % for NG, the specific gravity increases. This is probably due to the high polymer concentration, causing the clay surface to be saturated with polymer. Because of this, excess polymers could have remained in solution, probably causing the viscosity of the water to increase.

The presence of Na-CMC in the bentonite causes a dispersed structure, which creates flow paths that are more irregular. This will result in better hydraulic performance for the modified material. Taking the latter into account, it can be concluded that a polymer concentration between 8 % and 10 % should lead to an optimal hydraulic performance.

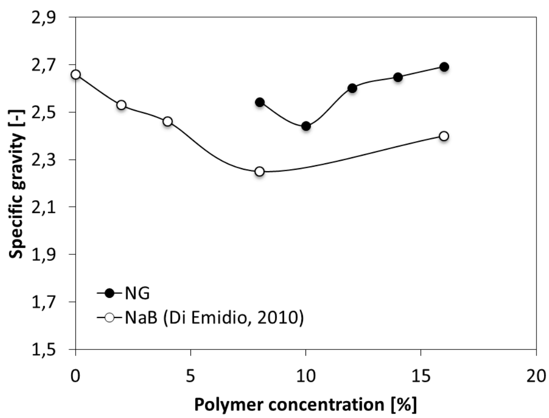

Figure 1. Specific gravity of sodium bentonite with increasing polymer dosage.

4.2 *Swell index and swell pressure test*

Figure 2 shows that the swell for the same solution concentration increases with increasing polymer concentration, regardless of the type of bentonite that is used. This effect is very clear for permeation with deionized water and up to a concentration of 0.001 M $CaCl_2$. Conversely, this effect starts to diminish at higher concentrations, which could be explained with the Gouy-Chapman theory of the DDL. It states that an increase in concentration leads to a smaller thickness of DDL, decreasing the amount of attracted water and affecting the swelling performance. Interestingly, CaB+16 % has a swelling potential which is similar or higher compared to NG+0 % and higher compared to SACaB+0 %. This means that the HYPER clay treatment on Ca-bentonites has an equal or better swelling potential to that of sodium and sodium-activated bentonite.

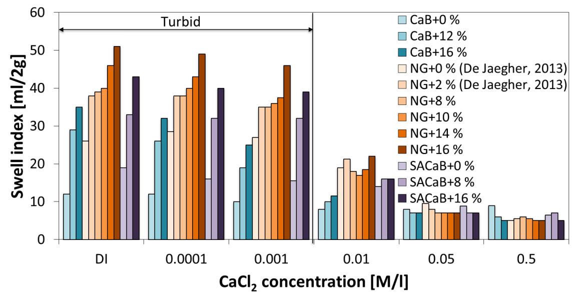

Figure 2. Swell index results of tested materials with various polymer concentrations.

As it was observed for the swell index values for DI, 0.0001, and 0.001 M $CaCl_2$, the bentonite caused turbidity in the test tubes. This is due to the low cation concentration in the permeating fluid allowing the DDL to resist collapse. The forces induced by the DDL between the clay platelets probably do not permit the clay particles to bond with one another, making them stay in suspension. The negatively charged polymer in between the clay particles intensified this effect. The turbidity causes the swell index to be an underestimation of the true value.

Swell index values were compared to those obtained by Seurynck (2012) in Figure 3. Interestingly enough, for DI and $CaCl_2$ concentrations ranging from 0.0001 to 0.01 M, the swell index values differ significantly. The method of sample preparation and the base bentonites were the same as in this study, but the configuration of the used Na-CMC was different. Seurynck (2012) used a powdered configuration, while this study used a granular configuration. A powdered configuration has a larger surface area, likely improving its adsorption to the clay surface. In this way, a more homogeneous distribution can be created, increasing the swelling performance significantly. From these results it can be concluded that a powdered Na-CMC should be used in future research, as it proves to have a higher swelling ability.

As expected, Figure 4 shows that Na-CMC treatment causes an increase in swell pressure performance compared to that of an untreated bentonite. Additionally, an increase in polymer dosage causes a higher swell pressure. It can also be seen that HYPER clay treatment of CaB has a beneficial effect compared to untreated SACaB. Two replicates of CaB+16 % samples give approximately the same swell pressure. Additionally, a CaB+16 % special sample was made. This sample contains 35.34g of clay and an additional 16 % polymer. The objective of this sample was to see whether there was a difference between the usual sample preparation (35.34g of clay, which includes the polymer) and the special sample preparation. As it can be seen from Figure 4, the special sample swell pressure is rather low. This is caused by the higher weight of the sample, causing the sample height to be higher, hence, the initial porosity to be higher as well. This is important as it will cause the swell pressure to be lower, due to the larger amount of voids that have to be filled before expansion can start.

Comparing values of Figure 4 with Figure 5 it was seen that the swell pressure for the same material was lower in this study compared to that in Di Emidio et al. (2013). This was caused by the swell pressure setup. The heights of the samples in this research were higher, which causes a higher initial porosity, resulting in a lower swelling pressure.

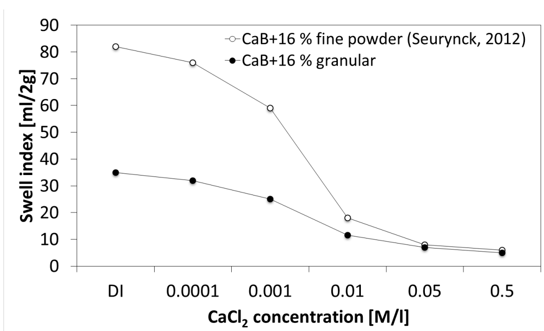

Figure 3. Powdered versus granular polymer

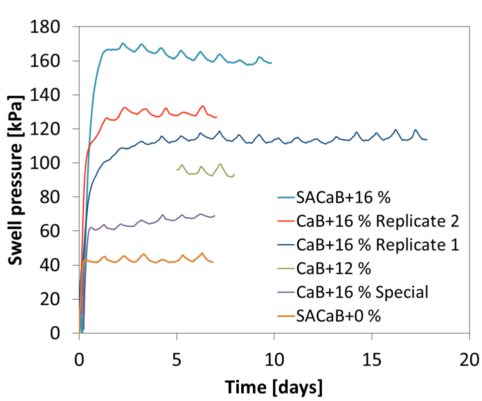

Figure 4. Swell pressure of calcium and sodium activated bentonite with varying polymer concentrations

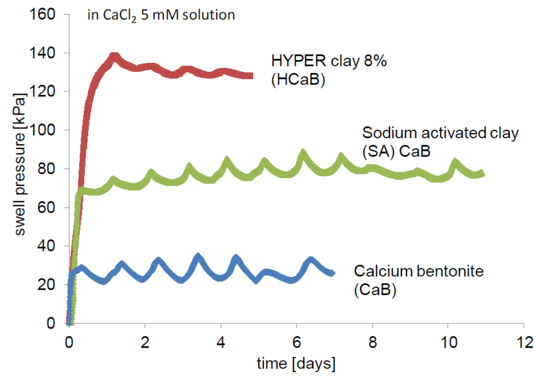

Figure 5. Swell pressure of calcium and sodium activated bentonite as well as HYPER clay (Di Emidio et al. 2013)

4.3 *Hydraulic conductivity*

As it can be seen from Figure 6, HYPER clay treatment of CaB decreases the hydraulic conductivity to 0.5 M $CaCl_2$ solution of about one order of magnitude compared to that of the untreated bentonite. The SACaB+16 % has a lower hydraulic conductivity compared to SACaB+0 %, which shows the beneficial effect of the HYPER clay treatment method. Additionally, the values for the special specimen are very similar to that of the CaB+16 % sample. This is probably due, as mentioned earlier in this paper, to the initial porosity of the special specimen being higher, causing a higher porosity.

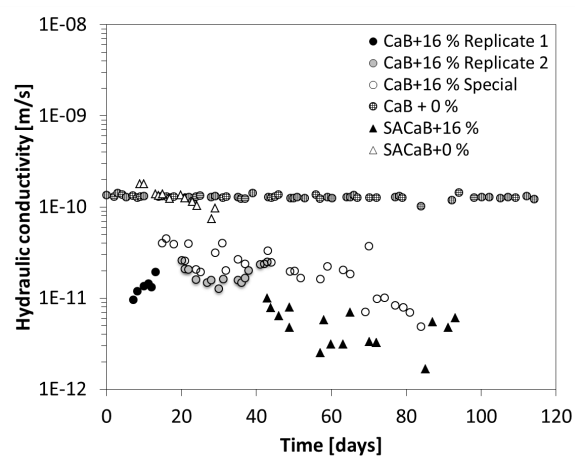

Figure 6. Hydraulic conductivity to 0.5 M $CaCl_2$ solution of CaB and SACaB treated and untreated with 16% of the polymer.

5 CONCLUSION

The specific gravity of NG showed that there is a possible optimum polymer adsorption at 10 % polymer concentration. The swelling ability of treated clays was quantified by means of standard swell index tests and swell pressure tests. Both showed that the treatment with the anionic polymer Na-CMC improved the swelling ability regardless of the type of bentonite that was used. The swell index test showed some limitations in its accuracy due to turbidity occurring at lower electrolyte concentrations. Nevertheless, it was seen that a powdered Na-CMC configuration resulted in a higher swelling ability compared to a granular configuration. On the other hand, swell pressure tests were not affected by the swell index limitations, leading to more reliable information on the swelling ability of clays. For $CaCl_2$ concentrations higher than 0.01 M, the swell index result was apparently not affected by the polymer treatment. Nevertheless, the HYPER clay treatment decreased the hydraulic conductivity to 0.5 M $CaCl_2$ of about 1-1.5 orders of magnitude compared to the untreated clay. The hydraulic conductivity to 0.5 M $CaCl_2$ solutions for the untreated calcium clay was indeed the highest. On the other hand, the hydraulic conductivity of the HYPER clay treated specimens were the lowest, due to the DDL being maintained thick in the long term even in presence of high concentration $CaCl_2$ solutions.

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